Transport fluctuation relations in interacting quantum pumps

New Journal of Physics
The open access journal at the forefront of physics

Deutsche Physikalische Gesellschaft DPG IOP Institute of Physics

PAPER • OPEN ACCESS

Transport fluctuation relations in interacting quantum pumps

To cite this article: Roman-Pascal Riwar and Janine Splettstoesser 2021 *New J. Phys.* **23** 013010

New Journal of Physics
The open access journal at the forefront of physics

Deutsche Physikalische Gesellschaft DPG
IOP Institute of Physics

Published in partnership with: Deutsche Physikalische Gesellschaft and the Institute of Physics

CrossMark

OPEN ACCESS

RECEIVED
28 October 2020

REVISED
12 December 2020

ACCEPTED FOR PUBLICATION
15 December 2020

PUBLISHED
20 January 2021

PAPER

Transport fluctuation relations in interacting quantum pumps

Roman-Pascal Riwar[1,*] **and Janine Splettstoesser**[2]

[1] Peter Grünberg Institute: Theoretical Nanoelectronics, Forschungszentrum Jülich, D-52425 Jülich, Germany
[2] Department of Microtechnology and Nanoscience, Chalmers University of Technology, S-41296 Göteborg, Sweden
[*] Author to whom any correspondence should be addressed.

E-mail: **r.riwar@fz-juelich.de**

Keywords: quantum transport, fluctuation relations, geometric phases, many-body interaction effects

Abstract

The understanding of out-of-equilibrium fluctuation relations in small open quantum systems has been a focal point of research in recent years. In particular, for systems with adiabatic time-dependent driving, it was shown that the fluctuation relations known from stationary systems do no longer apply due the geometric nature of the pumping current response. However, the precise physical interpretation of the corrected pumping fluctuation relations as well as the role of many-body interactions remained unexplored. Here, we study quantum systems with many-body interactions subject to slow time-dependent driving, and show that fluctuation relations of the charge current can in general not be formulated without taking into account the total energy current put into the system through the pumping process. Moreover, we show that this correction due to the input energy is nonzero only when Coulomb-interactions are present. Thus, fluctuation response relations offer an until now unrevealed opportunity to probe many-body correlations in quantum systems. We demonstrate our general findings at the concrete example of a single-level quantum dot model, and propose a scheme to measure the interaction-induced discrepancies from the stationary case.

1. Introduction

Fluctuation relations of quantum observables [1], and in particular their generalization to nonequilibrium driven quantum systems [2–16], are of fundamental importance to understand the second law of thermodynamics at the mesoscopic scale. The fluctuation relations themselves are formulated in terms of symmetries of the cumulant generating function. From these symmetries, a hierarchy of transport relations can be derived [3]. They relate different individual cumulants of charge and energy currents of different orders in the response to a chemical potential or temperature gradient, which is why they are now commonly referred to as fluctuation-*response* relations (FRR) [15]. From an application point of view, FRR can lead to powerful metrologic tools: for instance, the equilibrium fluctuation–dissipation theorem [1, 17] represents the centerpiece of Johnson thermometry (see reference [18] and references therein). From a fundamental perspective, the symmetries leading to the fluctuation relations might potentially become important for a better understanding of nonequilibrium topological phase transitions [19, 20].

In recent years, fluctuation relations for time-dependently driven systems have come to the focus of attention [8, 21–25]. This is timely, since experiments have advanced to the point that not only average charge currents, but also their fluctuations—the noise—could be measured accurately [26–32]. Even counting of charge on quantum-dot pumps has been realized [33, 34]. A main realization of this effort was, that fluctuation relations known from stationary systems do in general not simply extend to driven systems, due to the geometric nature of the system's response to the driving [8].

However, the previous theoretical works on fluctuation relations for quantum pumps [8, 22–25] have been for the most part carried out on a very general level in two respects. First of all, these works do not necessarily distinguish between charge or energy currents, and rather focus on the combined heat currents, or even completely generic 'place-holder' quantities without specification. Secondly, the additional step to derive the explicit FRR for specific cumulants has been omitted. For this reason, the mechanism underlying recently discovered deviations of the FRR in the presence of time-dependent driving compared to known stationary FRR in systems with strong many-body correlations [35, 36], remained unclear so far. Furthermore, we believe that this lack of specificity resulted in an incomplete understanding of the importance of many-body interactions in driven systems, first hinted at in reference [35].

In the present paper, we set up fluctuation relations and the resulting FRR for interacting quantum pumps, using the framework of full-counting statistics (FCS) for open quantum systems. We analyze in detail the resulting generalizations of the fluctuation dissipation theorem for the transported charge current and charge current noise. We focus in particular on adiabatic pumping, which is first and foremost of fundamental interest due to the geometric properties of the currents [24, 37–49]. Quantum pumps are furthermore of interest as controlled sources of a quantized current, see reference [50] (and references within).

In our analysis, we find that for pumping it is in general impossible to formulate FRR for charge transport only, without taking into account the energy input from the pump. Moreover, we can show that it is the specific geometric properties of the charge and energy transport, which are at the physical origin of deviations from stationary FRR. Namely, the explicit time-dependence of the eigenenergies of the quantum system breaks a global symmetry in the energy counting fields of the cumulant generating functions. As a consequence, the total energy input cannot be eliminated in the geometric response. Specifically, we find that the stationary fluctuation–dissipation theorem is violated due to the non-linear response of this pumping energy input. While heat transport and heat current fluctuations have been studied in time-dependently driven systems previously [51–55], this intricate connection between charge and energy transport had to our knowledge remained unknown so far.

Interestingly, we find in addition that the nonlinear-response character of the correction term has an important consequence: deviations from the standard stationary FRR occur only in the presence of many-body interactions. This, in particular, opens up the possibility to detect correlation effects via deviations from the standard FRR. Finally, we illustrate our general results at the specific example of a driven quantum dot with Coulomb interactions.

The paper is organized as follows. In section 2 we review the FCS framework for weakly coupled quantum systems and develop a systematic expansion for slowly driven pumps. Based on this framework, we derive a set of FRR in section 3, and explore the importance of many-body interactions. Finally, in section 4 we illustrate the derived FRR at the explicit model of a single-level interacting quantum dot and discuss their measurability.

2. FCS formalism for weakly coupled quantum pumps

We study the FCS of particle and energy currents through small quantum systems in the regime of weak tunnel coupling to the reservoirs [56] including a time-dependent drive [23]. In this section we review this framework, and state explicitly the assumptions under which it is valid. We then consistently expand the FCS in orders of the driving frequency, using a similar technique as for closed systems [57]. For the first order, the adiabatic-response (pumping) contribution, we recover the Sinitsyn–Nemenmann geometric phase [44]. In fact, the Sinitsyn–Nemenmann phase can be regarded as a geometric phase of the Landsberg type [40], where the necessary symmetry stems from the fact that the average pumped current is invariant with respect to a continuous recalibration of the charge meter, see references [47, 48]. The geometric nature of the pumping FCS is crucial for the properties of the cumulant generating function discussed in section 3.

2.1. Dynamics of the model system

We are interested in time-dependently driven quantum systems, exchanging particles and energy with reservoirs. We make the simplifying assumption that the tunnel coupling between the system and the reservoirs is weak, which is standard in the field [2–4, 6–9, 12, 14–16]. To study the effects of adiabatic pumping, we include a periodic driving of the system, which is slow with respect to the decay time of reservoir correlations. We will quantify these conditions in detail, in section 4.

We describe the dynamics of the system, using its reduced density matrix, represented in vector form $|P)$, which is obtained from the full density matrix by tracing out the reservoir degrees of freedom. The reduced density matrix fulfills a master equation

$$|\dot{P}) = \mathbf{W}(t)|P)\,. \tag{1}$$

Here, $\mathbf{W}(t)$ is a kernel, carrying all the transition rates of the stochastic processes that change the system state $|P)$. We here assume that the dynamics of the occupation probabilities of the system states does not couple to the dynamics of coherent superpositions, such that only the diagonal elements of the reduced density matrix matter[3]. In the weak coupling limit, which we are considering here, the rates can be computed through Fermi's golden rule of the *frozen* system where the time-dependence enters parametrically [60]. We otherwise keep a generic model for the following considerations and only specify a specific system in section 4.

Through the eigendecomposition of the kernel $\mathbf{W}$, we can understand the dynamics of the quantum system. Namely, the kernel can be decomposed into the form $\mathbf{W}(t) = \sum_k \lambda_k(t)|k(t))\,(k(t)|$, where the $|k)$ and $(k|$ are the right and left eigenvectors of $\mathbf{W}$, belonging to the eigenvalue λ_k. The notation is chosen such that any $|a)$ mathematically represents an operator, cast into vector form, whereas any $(a|$ represents a map from an operator to a scalar [61–63]. There is a zero mode $k = 0$, which expresses that for each t there exists a unique stationary state $|0)$ with $\lambda_0 = 0$. The corresponding (dual) left eigenvector $(0|$ is the trace operator, expressing the trace preserving property of $\mathbf{W}(t)$. The other eigenvalues and eigenmodes do not play a role in the long-time FCS of this slowly driven system, see equation (40) of reference [48].

Importantly, the system is coupled to several reservoirs, enumerated with α (and γ). In the weak coupling limit, the influence from the different reservoirs is additive $\mathbf{W} = \sum_\alpha \mathbf{W}_\alpha$. We assume microreversibility and local equilibrium for each reservoir, such that kernels have the symmetry [6] [4]

$$\mathbf{W}_\alpha^{\mathrm{T}}(t) = \mathrm{e}^{\beta_\alpha[\mu_\alpha\mathbf{n}-\mathbf{e}(t)]}\mathbf{W}_\alpha(t)\mathrm{e}^{-\beta_\alpha[\mu_\alpha\mathbf{n}-\mathbf{e}(t)]}. \tag{2}$$

where $\beta_\alpha = 1/(k_\mathrm{B}T_\alpha)$ and μ_α are, respectively, the inverse temperature and chemical potential of reservoir α. The particle number and energy superoperators associated to the local quantum system, $\mathbf{n} = \frac{1}{2}\{\hat{n},\cdot\}$ and $\mathbf{e} = \frac{1}{2}\{\hat{\epsilon},\cdot\}$, are defined via the anticommutators of the local particle number and energy operators, $\hat{n}$ and $\hat{\epsilon}$, respectively. They will be explicitly defined, when considering an explicit model system, see section 4.

We use the following notation throughout this work. The hat designates operators, which, when cast into vector form (in the superoperator context), are written in the round bra notation, $|\ldots)$. Superoperators are written in bold font, whereas regular fonts are used for scalars.

Finally, let us note that in principle, the chemical potential and temperature gradients may be time-dependent, too, since equation (2) is a time-local symmetry. However, in the remainder of this work, we keep μ_α and β_α constant in time in order to be able to clearly separate the response due to driving and the response due to chemical potential or temperature gradients. We will consider a time-dependent driving of the local system parameters, and the coupling amplitudes to the reservoirs, see section 4 for a concrete example.

2.2. Full counting statistics

We are not only interested in the mean dynamics of the system, but also in the FCS. Through integrating out the reservoir degrees of freedom, one has in principle lost all information about the transport statistics. However, one can keep track of the information of particle and energy transport by supplementing the kernels with counting fields (see reference [6] for a review),

$$\mathbf{W}(\{\chi_\alpha,\xi_\alpha\},t) = \sum_\alpha \mathrm{e}^{-\mathrm{i}\mathbf{e}(t)\xi_\alpha}\mathrm{e}^{-\mathrm{i}\mathbf{n}\chi_\alpha}\mathbf{W}_\alpha(t)\mathrm{e}^{\mathrm{i}\mathbf{n}\chi_\alpha}\mathrm{e}^{\mathrm{i}\mathbf{e}(t)\xi_\alpha}. \tag{3}$$

The counting fields χ_α and ξ_α keep track of the number of particles and the energy that enter reservoir α. Let us stress again, our main interest is in the charge currents, counted by χ_α. However, as we will show, the FRR for the charge current cannot be formulated and interpreted unless the energy currents are accounted for as well, such that we have to keep ξ_α.

Also for the kernel including counting fields, microreversibility imposes a symmetry, similar to equation (2), which can be expressed in terms of the counting fields through

$$\mathbf{W}^{\mathrm{T}}(\{\chi_\alpha,\xi_\alpha\},t) = \mathbf{W}(\{\mathrm{i}\beta_\alpha\mu_\alpha-\chi_\alpha,-\mathrm{i}\beta_\alpha-\xi_\alpha\},t). \tag{4}$$

This symmetry is of central importance for the formulation of fluctuation relations out of equilibrium [2–4, 6, 7, 9, 11, 13–16].

In order to obtain the particle and energy transport statistics, one needs to construct a so-called cumulant-generating function. One therefore starts from an arbitrary initial state $|P_0)$ at time t_in and then switches on the counting fields to measure the FCS until a certain time τ. Tracing over the remaining

[3] This widely used assumption is in particular valid for the driven single-level quantum dot, treated in section 4. A generalization to model systems where coherent dynamics are important can be done along the lines of e.g. [58] or [59].

[4] A generalization of the symmetries of the kernel to kernels connecting diagonal and off-diagonal elements of the density matrix can be envisaged along the lines of reference [12].

system degrees of freedom through application of $(0|$, one finds the cumulant generating function, $\mathcal{F}(\{\chi_\alpha, \xi_\alpha\}, \tau, t_{\rm in})$, from the evolution of the system in presence of the counting fields

$$\mathrm{e}^{\mathcal{F}(\{\chi_\alpha,\xi_\alpha\},\tau,t_{\rm in})(\tau-t_{\rm in})} = (0|\Pi(\{\chi_\alpha, \xi_\alpha\}, \tau, t_{\rm in})|P_0). \tag{5}$$

Here, we have introduced the propagator of the open system

$$\Pi(\{\chi_\alpha, \xi_\alpha\}, \tau, t_{\rm in}) = \mathcal{T}\mathrm{e}^{\int_{t_{\rm in}}^{\tau} \mathrm{d}t \mathbf{W}(\{\chi_\alpha,\xi_\alpha\},t)}. \tag{6}$$

The cumulants of the charge- and energy currents can now be computed by differentiating with respect to the counting fields of interest, and subsequently setting all counting fields to zero. For the remainder of this paper, we focus on the limit of very long measurement times $(\tau - t_{\rm in})/t_{\rm typ} \to \infty$, namely when $\tau - t_{\rm in}$ is much larger than typical system time scales $t_{\rm typ}$ given by the inverse of kernel eigenvalues $\lambda_k \neq 0$. Then $\mathcal{F}$ neither depends on τ nor $t_{\rm in}$ and provides the zero-frequency cumulants. In this case, the FCS does no longer depend on the initial state $|P_0)$, since for $\tau - t_{\rm in} \to \infty$, only the stationary state $|0)$ contributes to the transport statistics. Without loss of generality, we therefore set our initial state to be the stationary state in absence of the transport counting $|P_0) = |0(0))$. The zero-frequency cumulants of interest in the present paper, namely the average charge current into reservoir α and the related current–current correlations, are then defined as

$$I_\alpha \equiv I_\alpha(\tau - t_{\rm in} \to \infty) = -\mathrm{i} \lim_{\tau - t_{\rm in} \to \infty} \left[\partial_{\chi_\alpha} \mathcal{F}|_{\{\chi_\alpha,\xi_\alpha\}\to 0}\right], \tag{7}$$

as well as

$$S_{\alpha\gamma} \equiv S_{\alpha\gamma}(\tau - t_{\rm in} \to \infty) = - \lim_{\tau - t_{\rm in} \to \infty} \left[\partial_{\chi_\alpha}\partial_{\chi_\gamma} \mathcal{F}|_{\{\chi_\alpha,\xi_\alpha\}\to 0}\right]. \tag{8}$$

Importantly, and as foreshadowed already, also the energy current into reservoir α is found to play an important role in the present paper; it is defined via a derivative with respect to the energy counting field ξ_α,

$$I_{\mathrm{E},\alpha} \equiv I_{\mathrm{E},\alpha}(\tau - t_{\rm in} \to \infty) = -\mathrm{i} \lim_{\tau - t_{\rm in} \to \infty} \left[\partial_{\xi_\alpha} \mathcal{F}|_{\{\chi_\alpha,\xi_\alpha\}\to 0}\right]. \tag{9}$$

2.3. Adiabatic expansion

In order to find explicit expressions for the cumulant generating function, we now focus on the limit of slow driving as previously considered in [25, 45]. This means that the time-scale $\tau_0 = 2\pi/\Omega$, related to the inverse of the driving frequency Ω, is large with respect to the time scale on which the system states vary. In this adiabatic limit, the cumulant generating function $\mathcal{F}$ can be expanded in orders of the driving parameters, see appendix A. We evaluate it up to first order in the small driving parameter, $\mathcal{F} \approx \mathcal{F}^{(0)} + \mathcal{F}^{(1)}$. The zeroth-order, instantaneous contribution is given by[5]

$$\mathcal{F}^{(0)}(\{\chi_\alpha, \xi_\alpha\}) = \int_0^{\tau_0} \frac{\mathrm{d}t}{\tau_0} \lambda_0(\{\chi_\alpha, \xi_\alpha\}). \tag{10}$$

This is just a time-averaged version of the FCS which appear for a system without time-dependent driving. On top of that, we find the pumping contribution

$$\mathcal{F}^{(1)}(\{\chi_\alpha, \xi_\alpha\}) = -\int_0^{\tau_0} \frac{\mathrm{d}t}{\tau_0} (0(\{\chi_\alpha, \xi_\alpha\})|\partial_t|0(\{\chi_\alpha, \xi_\alpha\})). \tag{11}$$

This reproduces the result derived in reference [45] and shows the clearly geometric property of the adiabatic pumping transport statistics. This has important consequences: first of all, it is the properties of the *eigenvectors* and not only of the eigenvalues that enter here. Second, due to the time-dependent driving of system energies $\mathbf{e}(t)$, it can already be expected that the time-derivative will lead to terms involving the energy counting field, see also equation (3).

3. Charge current response relations and interaction effects

Our main goal is the derivation and interpretation of the FRR for charge currents using the symmetries due to microreversibility [6] of the cumulant generating function in the counting fields. Importantly, we show, that the geometric nature of the pumping contribution forbids in general that the charge FRR can be formulated without the appearance of an additional contribution, which is associated to the energy current

[5] Note that for simplicity of notation, we omitted the explicit time arguments in the eigenvalues and eigenvectors of W.

provided by the external pumping fields. We find this extra term to be a consequence of a drive-induced breaking of gauge invariance with respect to the energy counting fields ξ_α. We then make the additional surprising observation that the presence or absence of the additional energy dissipation term hinges on the presence or absence, respectively, of many-body interactions. Interestingly, while distinct in terms of the physical origin, this fact falls in line with various other interaction-induced pumping effects, see references [64, 65].

3.1. Fluctuation relations

Importantly, due to the above introduced symmetries of $\mathbf{W}(\{\chi_\alpha, \xi_\alpha\})$ [see equation (4)], we can derive fluctuation relations for $\mathcal{F}$. Namely we find that the instantaneous cumulant generating function satisfies the same relations as for a system without driving,

$$\mathcal{F}^{(0)}(\{\chi_\alpha, \xi_\alpha\}) = \mathcal{F}^{(0)}(\{\mathrm{i}\beta_\alpha\mu_\alpha - \chi_\alpha, -\mathrm{i}\beta_\alpha - \xi_\alpha\}), \tag{12}$$

whereas the pumping contribution satisfies

$$\mathcal{F}^{(1)}(\{\chi_\alpha, \xi_\alpha\}) = -\mathcal{F}^{(1)}(\{\mathrm{i}\beta_\alpha\mu_\alpha - \chi_\alpha, -\mathrm{i}\beta_\alpha - \xi_\alpha\}). \tag{13}$$

The minus sign for the pumping cumulant generating function reflects the fact that in order to satisfy microreversibility, the direction of pumping transport has to be inverted, too. The result shown in equations (12) and (13) has in another form been found previously [25] (i.e., in the form of a generic counting field, which could in principle encompass either charge or energy, or another observable quantity). Contrary to reference [25] however, we explicitly introduce the separate counting fields of charge and energy currents. It is this explicit distinction, which allows a careful derivation and interpretation of charge current FRR, as we will present them in the following.

3.2. Gauge transformations and their relationship to current conservation

As already announced, the first crucial step is to consider global gauge transformations of the cumulant generating function with respect to the counting fields. Both the instantaneous ($i = 0$) and the pumping contribution ($i = 1$) to the cumulant generating function, $\mathcal{F}$, are invariant with respect to global shifts of the *particle* counting fields,

$$\mathcal{F}^{(i)}(\{\chi_\alpha + \delta\chi, \xi_\alpha\}) = \mathcal{F}^{(i)}(\{\chi_\alpha, \xi_\alpha\}). \tag{14}$$

This invariance reflects the conservation of charge currents, which is valid irrespective of the presence or absence of a time-dependent driving. When applied on the level of the individual cumulants, we can derive the important and well-known identities

$$\sum_\alpha I_\alpha^{(i)} = 0 \tag{15a}$$

$$\sum_\alpha S_{\alpha\gamma}^{(i)} = \sum_\gamma S_{\alpha\gamma}^{(i)} = 0. \tag{15b}$$

However, the instantaneous and pumping contributions behave fundamentally differently from equation (14) with respect to a similar gauge in the energy counting fields, a fact which will be central for the rest of the discussion. While the instantaneous contribution exhibits a similar gauge invariance for ξ,

$$\mathcal{F}^{(0)}(\{\chi_\alpha, \xi_\alpha + \delta\xi\}) = \mathcal{F}^{(0)}(\{\chi_\alpha, \xi_\alpha\}), \tag{16}$$

the pumping correction does not. On the contrary, here, we receive an extra term

$$\mathcal{F}^{(1)}(\{\chi_\alpha, \xi_\alpha + \delta\xi\}) = \mathcal{F}^{(1)}(\{\chi_\alpha, \xi_\alpha\}) - \mathrm{i}\delta\xi\, \mathcal{Q}^{(1)}(\{\chi_\alpha, \xi_\alpha\}), \tag{17}$$

where we defined

$$\mathcal{Q}^{(1)}(\{\chi_\alpha, \xi_\alpha\}) = -\int_0^{\tau_0} \frac{\mathrm{d}t}{\tau_0} (0(\{\chi_\alpha, \xi_\alpha\})|\dot{\mathbf{e}}|0(\{\chi_\alpha, \xi_\alpha\})). \tag{18}$$

Formally, the origin of this different gauge behaviour, equations (14), (16) and (17), can be found when considering the kernel $\mathbf{W}(\{\chi_\alpha, \xi_\alpha\})$. Constant global shifts of the counting fields result in unitary transformations of the kernel, $\mathbf{W}(\{\chi_\alpha + \delta\chi, \xi_\alpha\}) = \mathrm{e}^{-\mathrm{i}\mathbf{n}\delta\chi}\mathbf{W}(\{\chi_\alpha, \xi_\alpha\})\mathrm{e}^{\mathrm{i}\mathbf{n}\delta\chi}$ respectively $\mathbf{W}(\{\chi_\alpha, \xi_\alpha + \delta\xi\}) = \mathrm{e}^{-\mathrm{i}\mathbf{e}(t)\delta\xi}\mathbf{W}(\{\chi_\alpha, \xi_\alpha\})\mathrm{e}^{\mathrm{i}\mathbf{e}(t)\delta\xi}$. Consequently, the *eigenvalues* of $\mathbf{W}$ remain unchanged, hence the gauge invariance of $\mathcal{F}^{(0)}$. The *eigenvectors* however, are transformed through the unitary super-operators $\exp(\pm\mathrm{i}\mathbf{n}\delta\chi)$ and $\exp(\pm\mathrm{i}\mathbf{e}(t)\delta\xi)$. Crucially, while a global shift in the particle counting field results in a time-independent transformation, the shift in the energy counting fields produces a *time-dependent* transformation. Hence, because of the geometric form of $\mathcal{F}^{(1)}$, see equation (11), it is easy to realize that the latter shift can in general not be eliminated, but results in the extra term on the right-hand side of equation (17).

Moreover, the breaking of this global symmetry can also be understood in physical terms. Namely, starting from equation (16), and solving for $\mathcal{Q}$, it is possible to show that

$$-\,\mathrm{i}\mathcal{Q}^{(1)} = \sum_{\gamma} \partial_{\xi_\gamma} \mathcal{F}^{(1)}. \tag{19}$$

Consequently, when putting all counting fields to zero, we can directly relate this function to the energy current provided by the external pumping fields,

$$\mathcal{Q}^{(1)}(\{0,0\}) = -\int_0^{\tau_0} \frac{\mathrm{d}t}{\tau_0} (0|\dot{\mathbf{e}}|0) = -\sum_{\alpha} I^{(1)}_{\mathrm{E},\alpha} \equiv I^{(1)}_{\mathrm{E,pump}}. \tag{20}$$

This reflects the very simple fact that energy is in general not conserved in the presence of an external drive, and can instead be pumped into the system. Charge conservation on the other hand is guaranteed even in the presence of the drive, such that no similar term arises for shifts in χ.

Let us now proceed and study the FRR for the pumping *charge* currents. We therefore have to determine the degree to which charge and energy counting fields can be disentangled in the presence of driving. The above derived gauge considerations are instrumental for this. From now on, we set $\beta_\alpha = \beta$ to avoid that the effect of driving is obscured. For the instantaneous contribution, equation (12), the energy counting fields can then just be set to zero (no counting of energy currents), and the global shift $-\mathrm{i}\beta$ appearing in the energy counting field argument can be gauged away [due to equation (16)]. This simply gives rise to

$$\mathcal{F}^{(0)}(\{\chi_\alpha\}) = \mathcal{F}^{(0)}(\{\mathrm{i}\beta\mu_\alpha - \chi_\alpha\}). \tag{21}$$

This is the time-averaged version of the well-known fluctuation relations for time-independent systems [2, 3] (see also equation (10)). Based on the above discussion, it is obvious that we cannot achieve the same 'charge current-only' response relations for the pumping contribution, equation (13). Namely, one cannot get rid of the pumping energy input $I_{\mathrm{E,pump}}$, and instead receives

$$\mathcal{F}^{(1)}(\{\chi_\alpha\}) = -\mathcal{F}^{(1)}(\{\mathrm{i}\beta\mu_\alpha - \chi_\alpha\}) + \beta\, \mathcal{Q}^{(1)}(\{\mathrm{i}\beta\mu_\alpha - \chi_\alpha\}). \tag{22}$$

We note that equation (22) automatically implies that

$$\mathcal{Q}^{(1)}(\{\chi_\alpha\}) = \mathcal{Q}^{(1)}(\{\mathrm{i}\beta\mu_\alpha - \chi_\alpha\}), \tag{23}$$

which can be seen by replacing $\chi_\alpha \to \mathrm{i}\beta\mu_\alpha - \chi_\alpha$ in equation (22). This means that remarkably, $\mathcal{Q}^{(1)}$ itself *does* satisfy a fluctuation relation quite similar to the instantaneous $\mathcal{F}^{(0)}$.

Equation (22) is one of the central results of this paper. We conclude that it is in general impossible to derive FRR for the charge pumping contribution, which involve only charge currents. The energy currents always enter due to the presence of the term $\mathcal{Q}^{(1)}$. We will see in the next section, that this contribution due to the pumping energy input is omnipresent when expressing the pumping fluctuation relations in terms of the individual cumulants, that is, the response relations.

3.3. FRR for charge pumping

The cumulant generating functions can now be expanded in terms of the counting fields and of the gradients in chemical potentials, in order to derive a resulting hierarchy of equations relating cumulants of different orders, yielding FRR. The derivation is detailed in appendix B.

In fact, the FRR for both instantaneous and first order in pumping can be expressed in a very compact form when introducing the total power provided by the external pumping field and the applied biases

$$J_{\mathrm{pump}} \equiv -\sum_{\alpha} \left(I_{\mathrm{E},\alpha} - \mu_\alpha I_\alpha\right) \equiv I_{\mathrm{E,pump}} + \sum_{\alpha} \mu_\alpha I_\alpha. \tag{24}$$

Then, we find both for the instantaneous ($i = 0$) and for the adiabatic-response contribution ($i = 1$),

$$0 = \left.\frac{\partial J^{(i)}_{\mathrm{pump}}}{\partial \mu_\alpha}\right|_{\{\mu_\alpha\}\to\mu} \tag{25}$$

$$S^{(i)}_{\alpha\gamma}\big|_{\{\mu_\alpha\}\to\mu} = k_{\mathrm{B}}T \left.\frac{\partial^2 J^{(i)}_{\mathrm{pump}}}{\partial\mu_\alpha \partial\mu_\gamma}\right|_{\{\mu_\alpha\}\to\mu}. \tag{26}$$

However, when expressed in terms of the power, the subtly different behavior of the instantaneous and pumping fluctuation relations is not clearly visible. We therefore from now on distinguish explicitly charge and energy currents, and discuss the resulting relations in detail.

For the instantaneous order, we get a time-averaged version of the response relations found also in reference [2, 3]. The lowest order relations read

$$I_\alpha^{(0)}\big|_{\{\mu_\alpha\}\to\mu} = 0 \tag{27}$$

$$S_{\alpha\gamma}^{(0)}\big|_{\{\mu_\alpha\}\to\mu} = k_\mathrm{B}T\left(\frac{\partial I_\alpha^{(0)}}{\partial\mu_\gamma}+\frac{\partial I_\gamma^{(0)}}{\partial\mu_\alpha}\right)\bigg|_{\{\mu_\alpha\}\to\mu}. \tag{28}$$

The first relation states that the time-averaged instantaneous currents must be zero in the absence of gradients in the chemical potentials, as expected. The second relation is a time-averaged version of the famous fluctuation–dissipation theorem.

As for the adiabatic-response, i.e. the pumping contribution, we find a first relation of the form

$$0 = I_{\mathrm{E,pump}}^{(1)}\Big|_{\{\mu_\alpha\}\to\mu}, \tag{29}$$

stating very simply that the pumping energy input is zero in the absence of chemical potential differences. This relation has no equivalent (or would be trivial) in the instantaneous order. It can be interpreted as follows. While one can induce a pumping current in the absence of chemical potential gradients (see also the FRR below), the electrons will be shuffled from one reservoir to the other, without putting in or extracting work. In addition, we find

$$I_\alpha^{(1)}\big|_{\{\mu_\alpha\}\to\mu} = \frac{\partial I_{\mathrm{E,pump}}^{(1)}}{\partial\mu_\alpha}\bigg|_{\{\mu_\alpha\}\to\mu} \tag{30}$$

$$S_{\alpha\gamma}^{(1)}\big|_{\{\mu_\alpha\}\to\mu} = k_\mathrm{B}T\left(\frac{\partial I_\alpha^{(1)}}{\partial\mu_\gamma}+\frac{\partial I_\gamma^{(1)}}{\partial\mu_\alpha}-\frac{\partial^2 I_{\mathrm{E,pump}}^{(1)}}{\partial\mu_\alpha\partial\mu_\gamma}\right)\bigg|_{\{\mu_\alpha\}\to\mu}. \tag{31}$$

Here, we see explicitly, what we have already indicated in the previous section, in equation (22), namely that the pumping energy input necessarily appears when attempting to formulate relations between different charge current cumulants. This seems to have been overlooked so far. While it is a well-known result (in many different physical regimes) that the pumping current can be non-zero even in the absence of a voltage bias [64, 66–69] we here show, that this nonzero pumping current can be related to the linear response of the energy current provided by the external pumping fields, $I_{\mathrm{E,pump}}$.

In equation (31), we recover a second FRR for pumping. Namely, while the instantaneous relation for the current noise, equation (28), satisfies a stationary (equilibrium) fluctuation dissipation theorem, this simple form cannot be extended to the pumping noise. Instead, it receives a correction due to the nonlinear response of $I_{\mathrm{E,pump}}$. A deviation from the fluctuation–dissipation theorem for the pumping noise has already been noted by us in reference [35] for a concrete single-level quantum dot model. In other works it has been implied that the stationary FDT does not extend to first order due to the geometric nature of the pumping cumulant generating function [8, 25]. However, in neither of these works the deviation from the stationary FDT has been explicitly identified or interpreted. Here, we find its explicit physical meaning as the nonlinear response of the pumping energy input.

3.4. The importance of charge conservation and many-body interactions

In this section, we show under which conditions the deviations from the standard FRR stemming from nonlinearities in the pumping energy input play a role.

The term, which we want to analyze in more detail here is the nonlinearity in the pumping energy input

$$\frac{\partial^2 I_{\mathrm{E,pump}}^{(1)}}{\partial\mu_\alpha\partial\mu_\gamma}\bigg|_{\{\mu_\alpha\}\to\mu}. \tag{32}$$

We start by considering the cross correlations, namely $\alpha\neq\gamma$. One immediately notices then that this term is nonzero only, if at least one of the summands in $I_{\mathrm{E,pump}}^{(1)}$ depends on the electrochemical potential of *two different* reservoirs. However, it is well known that in systems where many-body interactions are negligible the pumping charge and energy currents can be written in terms of incoming and outgoing effective distributions, where the different electrochemical potentials contribute to different summands, see e.g. equations (9) and (12) of reference [70]. This results in

$$\left.\frac{\partial^2 I^{(1)}_{\mathrm{E,pump}}}{\partial\mu_\alpha\partial\mu_\gamma}\right|_{\{\mu_\alpha\}\to\mu} \xrightarrow{\text{no interactions}} 0. \tag{33}$$

Crucially, we thus find that an equilibrium version of the FRR can be recovered for vanishing interactions. This is a further main result of this work. The deviation from the stationary FDT for the pumping response constitutes a purely interaction-induced effect, which is expected to be directly measurable in a noise or counting experiment. Moreover, this is a significant generalization of the result found in reference [35], where the recovery of the FRR for the pumping noise was found for the special case of a single-level quantum dot, including only two contacts. Here, the proof encompasses the broad class of generic quantum systems, weakly coupled to an arbitrary number of reservoirs, see section 2.

We now show how the above extends to autocorrelations via current conservation. Using equation (15a), one directly observes that the autocorrelations can be expressed as

$$S^{(1)}_{\alpha\alpha}\big|_{\{\mu_\alpha\}\to\mu} = -\sum_{\gamma\neq\alpha} S^{(1)}_{\alpha\gamma}\big|_{\{\mu_\alpha\}\to\mu}. \tag{34}$$

The constraint given in equation (34) can be rewritten using equation (31), leading to

$$\sum_\gamma \left.\frac{\partial I^{(1)}_\alpha}{\partial\mu_\gamma}\right|_{\{\mu_\alpha\}\to\mu} = \sum_\gamma \left.\frac{\partial^2 I^{(1)}_{\mathrm{E,pump}}}{\partial\mu_\alpha\partial\mu_\gamma}\right|_{\{\mu_\alpha\}\to\mu}. \tag{35}$$

Note, that the left-hand side of this equality is the sum over the adiabatic-response corrections to the charge conductances. In a non-driven system, this sum has to always equal zero. However, since the driving fields induce pumping charge currents, the extra term on the right-hand side of equation (35) appears, thereby constituting a generalization of the conductance sums for time-dependently driven systems.

Equation (35) can be rewritten as

$$\partial_\mu\left(I^{(1)}_\alpha\big|_{\{\mu_\alpha\}\to\mu}\right) = \partial_\mu\left(\left.\frac{\partial I^{(1)}_{\mathrm{E,pump}}}{\partial\mu_\alpha}\right|_{\{\mu_\alpha\}\to\mu}\right). \tag{36}$$

This shows that the correction of the sum of conductances can be derived from equation (30), by means of the derivative with respect to μ. Hence, the additional constraints for the auto-correlations are already fixed by the FRR for currents, equation (30).

4. Explicit model and the role of interactions

We have understood on a general level, how pumping modifies the FRR. The microreversibility relation, equation (4), corresponds to a gauge transformation, which leads to an extra term related to the energy current provided by the external pumping fields. We now consider an explicit quantum dot pump model to examine the properties of the fluctuation relations in detail. In particular we focus on how to probe the fluctuation relations in this experimentally relevant system, and illuminate the special role of electron-electron interactions.

4.1. FRR for a single-level quantum-dot pump

We consider a quantum dot model with a single, spin-degenerate level, tunnel-coupled to two reservoirs which may be at a different chemical potential, see figure 1(a). We have analyzed the charge pumping noise of this model in reference [35], and found that a deviation from the stationary fluctuation dissipation theorem occurs due to the time-dependent driving. Here, we identify the concrete physical mechanism for this deviation based on the pumping fluctuation relations presented in the previous section.

The total Hamiltonian is of the form

$$\hat{H} = \hat{H}_{\mathrm{QD}}(t) + \hat{H}_{\mathrm{T}}(t) + \sum_{\alpha=\mathrm{L,R}} \hat{H}_\alpha. \tag{37}$$

The local quantum dot Hamiltonian can be expressed as

$$\hat{H}_{\mathrm{QD}} = \sum_\sigma \epsilon(t)\hat{d}^\dagger_\sigma\hat{d}_\sigma + \frac{U}{2}\hat{n}\left(\hat{n}-1\right) \equiv \hat{\epsilon}. \tag{38}$$

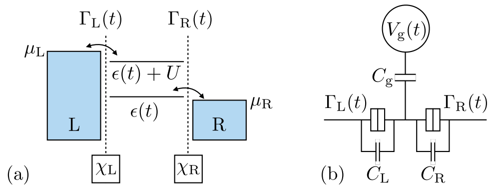

Figure 1. (a) Energy diagram of the single-level quantum dot. The quantum dot level has a time-dependent energy $\epsilon(t)$ and is tunnel coupled to a left (L) and right lead (R), with respective tunneling rates $\Gamma_{L,R}(t)$. The reservoirs may have a chemical potential difference, $\mu_L \neq \mu_R$. The charge counting fields $\chi_{L,R}$ are indicated at the tunnel barriers. (b) The circuit picture of the single-level quantum dot, valid when assuming geometric capacitances.

Here, the operators $\hat{d}_\sigma^{(\dagger)}$ annihilate (create) an electron with spin σ and energy $\epsilon(t)$. The parameter U expresses the magnitude of the onsite Coulomb interaction, which is sensitive to the electron occupation number $\hat{n} = \sum_\sigma \hat{d}_\sigma^\dagger \hat{d}_\sigma$. As indicated, the quantum dot Hamiltonian corresponds to the local energy operator $\hat{\epsilon}$. Out of these two operators, one can construct the explicit superoperators $\mathbf{n}$ and $\mathbf{e}$, as introduced in section 2. Note that while U may be time-dependent, too (as we will discuss later) we here keep it constant, in order to clearly distinguish between the interacting and non-interacting regimes. The Hamiltonians of the reservoirs are given as

$$\hat{H}_\alpha = \sum_{k\sigma} \epsilon_k \hat{c}_{\alpha k\sigma}^\dagger \hat{c}_{\alpha k\sigma}, \tag{39}$$

where likewise the operators $\hat{c}_{\alpha k\sigma}^{(\dagger)}$ annihilate (create) an electron in reservoir α, with momentum k and spin σ, as well as energy ϵ_k. Finally, the tunneling Hamiltonian is

$$\hat{H}_T(t) = \sum_{\alpha k\sigma} \gamma_\alpha(t) \hat{c}_{\alpha k\sigma}^\dagger \hat{d}_\sigma + \text{h.c.}, \tag{40}$$

where $\gamma_\alpha(t)$ denotes the time-dependent tunneling amplitude. All the time-dependent parameters shall be subject to a periodic driving, $x(t+\tau_0) = x(t)$, where the pumping period is $\tau_0 = 2\pi/\Omega$ with the driving frequency Ω. To simplify the notation, we will from now on omit the brackets (t) for these parameters, and for any further quantities depending on them.

We now present the rate equation of the quantum dot system, in presence of the tunnel coupling. In the sequential tunneling limit, valid for small tunnel couplings, $\Gamma_\alpha \ll k_B T$, with $\Gamma_\alpha = 2\pi\rho_\alpha |\gamma_\alpha|^2$, the dynamics of the system are of the form of equation (1) introduced in section 2. For the here considered system, the reduced density matrix in vector form reads $|P) = (P_0, P_\uparrow, P_\downarrow, P_2)^T$, where only the diagonal elements are relevant[6]. They contain the occupation probabilities of the quantum dot with the probability of the quantum dot being empty P_0, singly occupied with either an $\uparrow$ or $\downarrow$ electron $P_{\uparrow,\downarrow}$, or doubly occupied P_2. In the following, we focus on a regime of adiabatic driving[7], $\Omega \ll \Gamma_\alpha$, where the expansion presented in appendix A applies[8] and where we compute the dynamics following references [60, 64].

The kernel describing the time evolution can be given as $\mathbf{W} = \sum_{\alpha=L,R} \mathbf{W}_\alpha$ and,

$$\mathbf{W}_\alpha = \Gamma_\alpha \begin{pmatrix} -2f_\alpha & \bar{f}_\alpha & \bar{f}_\alpha & 0 \\ f_\alpha & -\bar{f}_\alpha - f_\alpha^U & 0 & \bar{f}_\alpha^U \\ f_\alpha & 0 & -\bar{f}_\alpha - f_\alpha^U & \bar{f}_\alpha^U \\ 0 & f_\alpha^U & f_\alpha^U & -2\bar{f}_\alpha^U \end{pmatrix} \tag{41}$$

with $f_\alpha = f(\epsilon - \mu_\alpha)$, $f_\alpha^U = f(\epsilon + U - \mu_\alpha)$, and $\bar{f} = 1 - f$, where $f(E) = 1/(e^{E/k_B T} + 1)$ is the Fermi function. Likewise, we can write the particle number and energy superoperators as $\mathbf{n} = \text{diag}(0, 1, 1, 2)$ and $\mathbf{e} = \text{diag}(0, \epsilon, \epsilon, 2\epsilon + U)$. With the above ingredients, we can now construct the kernel including the particle and energy current counting fields, according to equation (3). Thus we find the FCS of a quantum dot pump, and can therefore test the fluctuation relations elaborated in the previous sections. In particular, the kernels $\mathbf{W}_\alpha$ fulfill the symmetry relation of equation (2), and consequently equation (3) follows from that.

[6] For the model considered here, this is valid as long as the contacts are normal metals, guaranteeing charge and spin conservation.
[7] Depending on the driving amplitudes, this condition should be generalized to $\delta\varepsilon\Omega/\Gamma k_B T \ll 1$ with the amplitude of the time-dependent energy level $\delta\varepsilon$, see reference [65].
[8] In principle, this allows us to consider the dynamics even for driving faster than the tunneling dynamics, as long as the driving occurs on time scales slower than $(k_B T)^{-1}$

The analytic expressions for the pumping current and pumping noise for this single-level quantum-dot pump have already been computed in reference [35]. For the purpose of this paper, we need the expression for the current for arbitrary chemical potentials (because we will have to compute the conductance below), whereas the noise is only required at $\mu_{\mathrm{L}} = \mu_{\mathrm{R}} = \mu$. The respective expressions are

$$I_{\mathrm{L}}^{(1)}\Big|_{\{\mu_\alpha\}\to\mu} = -\frac{1}{2}\int_0^{\tau_0}\frac{\mathrm{d}t}{\tau_0}\frac{\lambda_{\mathrm{c,L}}-\lambda_{\mathrm{c,R}}}{\lambda_{\mathrm{c}}}\partial_t\langle\hat{n}\rangle \tag{42}$$

$$S_{\mathrm{LR}}^{(1)}\Big|_{\{\mu_\alpha\}\to\mu} = \int_0^{\tau_0}\frac{\mathrm{d}t}{\tau_0}\frac{\Gamma_{\mathrm{L}}\Gamma_{\mathrm{R}}}{\Gamma^2}\partial_t\Delta n, \tag{43}$$

where $\lambda_{\mathrm{c},\alpha} = \Gamma_\alpha\left(1+f_\alpha-f_\alpha^U\right)$ is the charge relaxation rate due to coupling to lead α, and $\lambda_{\mathrm{c}} = \sum_\alpha\lambda_{\mathrm{c},\alpha}$ is the total charge relaxation rate. While $\lambda_{\mathrm{c},\alpha}$ are eigenvalues of the respective W_α, the sum λ_{c} is an eigenvalue of the total W. The dot occupation expectation value is defined as $\langle\hat{n}\rangle = (0|\mathbf{n}|0)$. For arbitrary chemical potentials it reads

$$\langle\hat{n}\rangle = 2\left(\frac{\Gamma_{\mathrm{L}}}{\lambda_{\mathrm{c}}}f_{\mathrm{L}} + \frac{\Gamma_{\mathrm{R}}}{\lambda_{\mathrm{c}}}f_{\mathrm{R}}\right). \tag{44}$$

Its fluctuations are defined as $\Delta n = \langle\hat{n}^2\rangle - \langle\hat{n}\rangle^2 = (0|\mathbf{n}^2|0) - (0|\mathbf{n}|0)^2$. For the noise expression equation (43), we only need the limit $\mu_{\mathrm{L}} = \mu_{\mathrm{R}} = \mu$, which is

$$\Delta n = -k_{\mathrm{B}}T\partial_\epsilon\langle\hat{n}\rangle. \tag{45}$$

These expressions yield an explicit form for the deviations from the standard stationary FRR for a single-level quantum-dot pump, which we could compactly write as [35]

$$\begin{aligned} S_{\mathrm{LR}}^{(1)}\Big|_{\{\mu_\alpha\}\to\mu} - k_{\mathrm{B}}T\left(\frac{\partial I_{\mathrm{L}}^{(1)}}{\partial\mu_{\mathrm{R}}}+\frac{\partial I_{\mathrm{R}}^{(1)}}{\partial\mu_{\mathrm{L}}}\right)\Bigg|_{\{\mu_\alpha\}\to\mu} \\ = -2k_{\mathrm{B}}T\int_0^{\tau_0}\frac{\mathrm{d}t}{\tau_0}\frac{\Gamma_{\mathrm{L}}\Gamma_{\mathrm{R}}}{\Gamma^2}\frac{\partial_\epsilon\lambda_{\mathrm{c}}}{\lambda_{\mathrm{c}}}\partial_t\langle\hat{n}\rangle. \end{aligned} \tag{46}$$

We will now show that the term on the right-hand side fulfills the main results presented in III, namely that it (a) vanishes for negligible Coulomb interaction and (b) that it equals the nonlinear contributions to the energy-current provided by the external pumping fields.

(a) For $U = 0$ the charge relaxation rate λ_{c} simply equals the coupling constant Γ. It is hence constant with respect to the dot energy, $\partial_\epsilon\lambda_{\mathrm{c}} = 0$, and consequently the right-hand-side of equation (46) must be zero.

(b) Based on equation (20), we can express the energy current provided by the external pumping fields for the single-level quantum dot as,

$$I_{\mathrm{E,pump}}^{(1)} = -\int_0^{\tau_0}\frac{\mathrm{d}t}{\tau_0}(0|\dot{\mathbf{e}}|0) = -\int_0^{\tau_0}\frac{\mathrm{d}t}{\tau_0}\dot{\epsilon}\langle\hat{n}\rangle. \tag{47}$$

With straightforward algebra, one can now explicitly show that

$$\frac{\partial^2 I_{\mathrm{E,pump}}^{(1)}}{\partial\mu_{\mathrm{L}}\partial\mu_{\mathrm{R}}}\Bigg|_{\{\mu_\alpha\}\to\mu} = 2\int_0^{\tau_0}\frac{\mathrm{d}t}{\tau_0}\frac{\Gamma_{\mathrm{L}}\Gamma_{\mathrm{R}}}{\Gamma^2}\frac{\partial_\epsilon\lambda_{\mathrm{c}}}{\lambda_c}\partial_t\langle\hat{n}\rangle, \tag{48}$$

thus satisfying equation (31). In figure 2, we plot the respective quantities $S^{(1)} \equiv S_{\mathrm{LR}}^{(1)}$, $G^{(1)} \equiv \partial_{\mu_{\mathrm{R}}}I_{\mathrm{L}}^{(1)} + \partial_{\mu_{\mathrm{L}}}I_{\mathrm{R}}^{(1)}$, and $Q^{(1)} \equiv \partial_{\mu_{\mathrm{L}}}\partial_{\mu_{\mathrm{R}}}I_{\mathrm{E,pump}}^{(1)}$ as a function of the time-averaged energy level $\overline{\epsilon}$. For the noninteracting system (a), we recover the equilibrium fluctuation dissipation theorem, in spite of the presence of a nonequilibrium drive. As soon as finite interactions are present (b), the correction due to the quadratic response of the pumping energy input, $I_{\mathrm{E,pump}}^{(1)}$ is nonzero, such that the equilibrium fluctuation dissipation theorem is no longer applicable. The interaction-induced deviation from it is significant, as $I_{\mathrm{E,pump}}^{(1)}$ is visibly of the same order of magnitude as $\partial_{\mu_{\mathrm{R}}}I_{\mathrm{L}}^{(1)} + \partial_{\mu_{\mathrm{L}}}I_{\mathrm{R}}^{(1)}$.

4.2. Validation of fluctuation relations through charge currents only

Crucially, equation (47) indicates how the fluctuation relation in equation (46) could be experimentally verified. In general, one would need the information of both the current and current noise, as well as the total energy current provided by the external pumping fields. In particular the direct measurement of

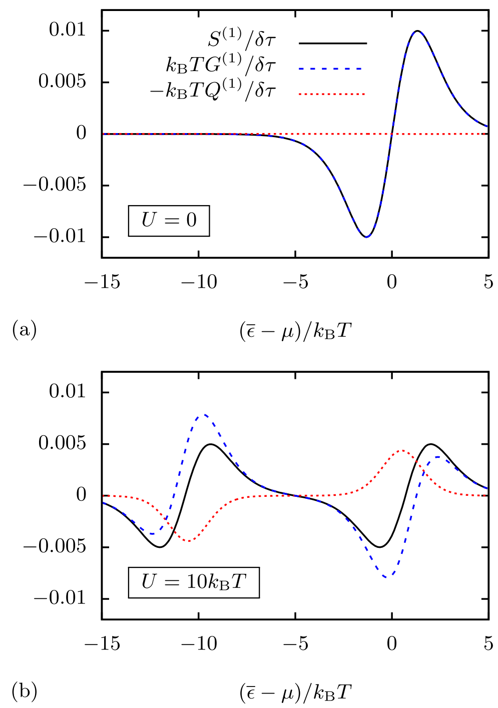

Figure 2. Pumping charge current noise $S^{(1)} \equiv S^{(1)}_{\mathrm{LR}}|_{\{\mu_\alpha\}\to\mu}$, conductance $G^{(1)} \equiv (\partial I^{(1)}_{\mathrm{L}}/\partial\mu_{\mathrm{R}} + \partial I^{(1)}_{\mathrm{R}}/\partial\mu_{\mathrm{L}})|_{\{\mu_\alpha\}\to\mu}$, and nonlinearity in the pumping energy input $Q^{(1)} \equiv \partial^2 I^{(1)}_{\mathrm{E,pump}}/\partial\mu_{\mathrm{L}}\partial\mu_{\mathrm{R}}|_{\{\mu_\alpha\}\to\mu}$, as a function of the average energy level $\bar{\epsilon}$. The interaction strength is (a) $U = 0$ and (b) $U = 10k_{\mathrm{B}}T$. We choose $\epsilon(t) = \bar{\epsilon} + \delta\epsilon\,\sin(\Omega t + \phi)$ and $\Gamma_{\mathrm{L}} = \overline{\Gamma}_{\mathrm{L}} + \delta\Gamma\,\sin(\Omega t)$ as the driving parameters, with $\phi = \pi/2$. The values of the parameters are $\overline{\Gamma}_{\mathrm{L}} = 0.65\overline{\Gamma}$, $\Gamma_{\mathrm{R}} = 0.35\overline{\Gamma}$, such that $\overline{\Gamma} = \overline{\Gamma}_{\mathrm{L}} + \Gamma_{\mathrm{R}}$. The driving parameters are $\delta\epsilon = 0.1k_{\mathrm{B}}T$, $\delta\Gamma = 0.1\overline{\Gamma}$. The quantities are plotted with respect to the rate $1/\delta\tau \equiv \Omega\delta\Gamma_{\mathrm{L}}\delta\epsilon/(\overline{\Gamma}k_{\mathrm{B}}T)$.

energy currents may be challenging, even if current progress on fast thermometry could open up new opportunities [71, 72].

However, equation (47) indicates that one could alternatively measure the quantum dot occupation number $\langle\hat{n}\rangle$ as a function of time, which can be done through a local charge detector such as a quantum point contact [33, 34]. Importantly, we note, that the time resolution need not be high. The measuring frequency ω (such that ω^{-1} can be regarded as a sampling time scale, that is, the detector resolution in time space) needs to be higher than the driving frequency Ω, but in the adiabatic-response limit, can still be smaller than the tunneling rate Γ_α.

The remaining quantity one needs to have access to is $\epsilon(t)$. In experiment, ϵ is typically driven through an external gate voltage. Resorting to a circuit picture, see figure 1(b), and assuming that the electrostatics of the system can be fully described by geometric capacitances, we can relate $\epsilon(t) = a\mathrm{e}V_{\mathrm{g}}(t) + \mathrm{const.}$, with the lever arm factor $a = C_{\mathrm{g}}/(C_{\mathrm{g}} + C_{\mathrm{L}} + C_{\mathrm{R}})$. Consequently, the correction on the right-hand side of equation (46) is accessible through the input function $V_{\mathrm{g}}(t)$, the output function $\langle\hat{n}\rangle(t)$ of the charge detector, and a single fitting parameter a.

Finally, we comment on the case where the Coulomb interaction is time-dependent, $U(t)$, and used as one of the pumping parameters [73]. Within the model of geometric capacitances, where $U = \mathrm{e}^2/(C_{\mathrm{g}} + C_{\mathrm{L}} + C_{\mathrm{R}})$, a time-dependent U can arise if the driving of the quantum dot changes the geometry, meaning that the capacitances themselves become time dependent. Also tunable effective Coulomb interaction has been realized [74]. Importantly, the general pumping fluctuation relations from section 3.1, see equation (31), encompass also such a time-dependent driving of U—only the explicit shape for the pumping energy input, equation (48), needs to be modified. This total energy current can then be decomposed into a charge and a parity contribution, as

$$I^{(1)}_{\mathrm{E,pump}} = -\int_0^{\tau_0}\frac{\mathrm{d}t}{\tau_0}\left[\left(\dot{\epsilon} + \frac{\dot{U}}{2}\right)\langle\hat{n}\rangle - \frac{\dot{U}}{2}\langle\hat{p}\rangle\right], \tag{49}$$

where $\hat{p} = \mathrm{e}^{\mathrm{i}\pi\hat{n}}$ is the parity operator. For this specific model, this yields the nonlinear relation $\hat{p} = 1 - 4\hat{n} + 2\hat{n}^2$. It is likewise accessible by a time-resolved charge detector. Instead of measuring the total charge, one here needs to keep track of the number of single electron tunneling events, and whether the resulting occupation is odd or even. The remaining task is to experimentally control the geometry of the device to the extent that reliable information of $U(t)$ as a function of the driving parameters can be found.

5. Conclusions

In this work, we have studied fluctuation relations of adiabatic quantum pumps. We have shown that due to the time-dependent driving, one cannot formulate fluctuation relations of charge currents without taking into account the energy current provided by the external pumping fields. This energy current necessarily enters due to the geometric nature of the pumping observables. Namely, the elimination of the energy counting fields comes with a time-dependent unitary transformation, to which the geometric response is sensitive.

As a result, the FRR of the quantum pump, obtained from derivatives of the cumulant generating function with respect to the counting fields, tie the charge current noise to the total power provided by the external driving. Interestingly, while deviations from the FRR valid for stationary systems can be expected due to pumping, we here find that they are fully induced by many-body interactions on the local quantum system.

We have concretely studied the FRR at the example of a single-level quantum dot with time-dependent energy-level and tunnel coupling. Even for this simple case the corrections to the FRR due to the pumping energy input are in general of the same order of magnitude as the pumping contributions to charge current and charge current noise, and are hence not negligible. Furthermore, we have sketched possible experimental strategies to verify our pumping fluctuation relations, even if energy currents cannot be measured.

As an outlook, it would be interesting to extend this study to finite-time FCS and the FRR for finite-frequency observables. This could establish connections between deviations from finite-frequency FRR due to strong interactions, discovered in reference [36] and first connections between finite frequency noise and energy currents established for noninteracting systems in reference [21].

Acknowledgments

We thank F Haupt, M R Wegewijs and M Esposito for fruitful discussions. J S acknowledges financial support by the Knut and Alice Wallenberg Foundation (via a Fellowship grant) and the Vetenskapsrådet, Swedish VR. R-P R acknowledges financial support by the German Federal Ministry of Education and Research within the funding program Photonic Research Germany under the Contract No. 13N14891.

Appendix A. Adiabatic expansion

Here, we briefly explain the adiabatic expansion for the cumulant generating function. As a matter of fact, in spite of dealing with a dissipative open quantum system, this expansion can be envisaged in close analogy to the case of closed Hamiltonian systems [57].

Exploiting the eigenvector decomposition of $\mathbf{W}(\{\chi_\alpha, \xi_\alpha\})$, we can expand the propagator in orders of the parameter $(k|\dot{k}')/(\lambda_k - \lambda_{k'}) \simeq \Omega/\Gamma$. The lowest, zeroth order term of this FCS propagator results in

$$\Pi(t,t') \approx \mathrm{e}^{\int_{t'}^{t} \mathrm{d}t_1 \left[\lambda_0(t_1) - (0|\dot{0})(t_1)\right]} |0(t))\,(0(t')|, \tag{A1}$$

which is valid for sufficiently long $t - t'$, where the terms with $k > 0$ will be exponentially suppressed.

Note that this zeroth order expansion of the FCS includes the first-order nonadiabatic correction of the transport equations, see references [47, 48].

Appendix B. Deriving FRR from fluctuation relations

Starting from the fluctuation relations in the main text, equations (21) and (22), we now derive the FRR given in equations (27)–(31). For this purpose, we set the counting fields χ_α and ξ_α to zero in equations (21) and (22). We receive

$$0 = \mathcal{F}^{(0)}\left(\{\mathrm{i}\beta\mu_\alpha\}\right), \tag{B1}$$

and

$$\beta I^{(1)}_{\mathrm{E,pump}} = \mathcal{F}^{(1)}\left(\{\mathrm{i}\beta\mu_\alpha\}\right), \tag{B2}$$

where for the second equation we used the identities from equations (23) and (20). As a next step, we expand both equations in a Taylor series in μ_α. While for $I_{\mathrm{E,pump}}$ this expansion is trivial, note that the quantities $\mathcal{F}^{(i)}$ do not only depend on μ_α through the remaining counting field arguments $\{\mathrm{i}\beta\mu_\alpha\}$, but they also explicitly depend on μ_α through the μ_α-dependence of the bare W (i.e., in the absence of the counting fields). Hence, the expansion up to second order in $\delta\mu_\alpha = \mu_\alpha - \mu$ provides us here with

$$\begin{aligned}\mathcal{F}^{(i)}\left(\{\mathrm{i}\beta\mu_\alpha\}\right) = &\sum_\alpha \delta\mu_\alpha \partial_{\mu_\alpha}\mathcal{F}\Big|_{\substack{\{\mu_\alpha\}\to\mu \\ \{\chi_\alpha\}\to 0}} \\ &+ \mathrm{i}\beta\sum_\alpha \delta\mu_\alpha \partial_{\chi_\alpha}\mathcal{F}^{(i)}\Big|_{\substack{\{\mu_\alpha\}\to\mu \\ \{\chi_\alpha\}\to 0}} \\ &+ \mathrm{i}\beta\sum_{\alpha\gamma} \delta\mu_\alpha\delta\mu_\gamma \partial_{\chi_\alpha}\partial_{\mu_\gamma}\mathcal{F}^{(i)}\Big|_{\substack{\{\mu_\alpha\}\to\mu \\ \{\chi_\alpha\}\to 0}} \\ &- \frac{1}{2}\beta^2\sum_{\alpha\gamma} \delta\mu_\alpha\delta\mu_\gamma \partial_{\chi_\alpha}\partial_{\chi_\gamma}\mathcal{F}^{(i)}\Big|_{\substack{\{\mu_\alpha\}\to\mu \\ \{\chi_\alpha\}\to 0}} \\ &+ \cdots. \end{aligned} \tag{B3}$$

Since equations (B1) and (B2) have to be satisfied for arbitrary values of the chemical potentials μ_α, we may collect terms with the same order in $\delta\mu_\alpha$ and demand that they fulfill equations (B1) and (B2) individually. Taking into account the definitions for the current and noise in equations (7) and (8), we arrive at equations (27)–(31).

Finally, let us point out that the relationship between the FRR in equations (27)–(31) and the more compact version in terms of the total pumping power, J_{pump}, see equations (25) and (26) can be seen as follows. Based on the definition of J_{pump} in equation (24), we see that when differentiating it with respect to μ_α, we get

$$\partial_{\mu_\alpha}J_{\mathrm{pump}} = \partial_{\mu_\alpha}I_{\mathrm{E,pump}} + \sum_\gamma \delta_{\alpha\gamma}I_\gamma + \sum_\gamma \mu_\gamma\partial_{\mu_\alpha}I_\gamma. \tag{B4}$$

Taking subsequently the limit of all $\mu_\alpha \to \mu$, we see that the second term on the right-hand side has to vanish due to current conservation, thus leading straight from equations (25) to (30). The very same principle applies to the second order derivative, relating equations (26) to (31), which we do not show explicitly.

ORCID iDs

Roman-Pascal Riwar https://orcid.org/0000-0002-0402-1090
Janine Splettstoesser https://orcid.org/0000-0003-1078-9490

References

[1] Callen H B and Welton T A 1951 Irreversibility and generalized noise *Phys. Rev.* **83** 34–40
[2] Esposito M and Mukamel S 2006 Fluctuation theorems for quantum master equations *Phys. Rev.* E **73** 046129
[3] Andrieux D and Gaspard P 2007 A fluctuation theorem for currents and non-linear response coefficients *J. Stat. Mech.* P02006
[4] Esposito M, Harbola U and Mukamel S 2007 Entropy fluctuation theorems in driven open systems: application to electron counting statistics *Phys. Rev.* E **76** 031132
[5] Förster H and Büttiker M 2008 Fluctuation relations without microreversibility in nonlinear transport *Phys. Rev. Lett.* **101** 136805
[6] Esposito M, Harbola U and Mukamel S 2009 Nonequilibrium fluctuations, fluctuation theorems, and counting statistics in quantum systems *Rev. Mod. Phys.* **81** 1665–702
[7] Utsumi Y, Golubev D S, Marthaler M, Saito K, Fujisawa T and Schön G 2010 Bidirectional single-electron counting and the fluctuation theorem *Phys. Rev.* B **81** 125331
[8] Ren J, Hänggi P and Li B 2010 Berry-phase-induced heat pumping and its impact on the fluctuation theorem *Phys. Rev. Lett.* **104** 170601
[9] Campisi M, Hänggi P and Talkner P 2011 Colloquium: quantum fluctuation relations: foundations and applications *Rev. Mod. Phys.* **83** 771–91
[10] Saira O-P, Yoon Y, Tanttu T, Möttönen M, Averin D V and Pekola J P 2012 Test of the Jarzynski and Crooks fluctuation relations in an electronic system *Phys. Rev. Lett.* **109** 180601
[11] Lippiello E, Baiesi M and Sarracino A 2014 Nonequilibrium fluctuation–dissipation theorem and heat production *Phys. Rev. Lett.* **112** 140602
[12] Ansari M H and Nazarov Y V 2015 Rényi entropy flows from quantum heat engines *Phys. Rev.* B **91** 104303
[13] Borrelli M, Koski J V, Maniscalco S and Pekola J P 2015 Fluctuation relations for driven coupled classical two-level systems with incomplete measurements *Phys. Rev.* E **91** 012145
[14] Cuetara G B, Engel A and Esposito M 2015 Stochastic thermodynamics of rapidly driven systems *New J. Phys.* **17** 055002
[15] Altaner B, Polettini M and Esposito M 2016 Fluctuation–dissipation relations far from equilibrium *Phys. Rev. Lett.* **117** 180601

[16] Benito M, Niklas M and Kohler S 2016 Full-counting statistics of time-dependent conductors *Phys. Rev.* B **94** 195433
[17] Kubo R 1966 The fluctuation–dissipation theorem *Rep. Prog. Phys.* **29** 255–84
[18] White D R *et al* 1996 The status of Johnson noise thermometry *Metrologia* **33** 325–35
[19] Ren J and Sinitsyn N A 2013 Braid group and topological phase transitions in nonequilibrium stochastic dynamics *Phys. Rev.* E **87** 050101
[20] Riwar R-P 2019 Fractional charges in conventional sequential electron tunneling *Phys. Rev.* B **100** 245416
[21] Moskalets M and Büttiker M 2009 Heat production and current noise for single- and double-cavity quantum capacitors *Phys. Rev.* B **80** 081302
[22] Bulnes Cuetara G, Esposito M and Imparato A 2014 Exact fluctuation theorem without ensemble quantities *Phys. Rev.* E **89** 052119
[23] Croy A and Saalmann U 2016 Full counting statistics of a nonadiabatic electron pump *Phys. Rev.* B **93** 165428
[24] Watanabe K L and Hayakawa H 2017 Geometric fluctuation theorem for a spin-boson system *Phys. Rev.* E **96** 022118
[25] Hino Y and Hayakawa H 2020 Fluctuation relations for adiabatic pumping *Phys. Rev.* E **102** 012115
[26] Flindt C, Fricke C, Hohls F, Novotný T, Netocny K, Brandes T and Haug R J 2009 Universal oscillations in counting statistics *Proc. Natl Acad. Sci.* **106** 10116–9
[27] Parmentier F D, Bocquillon E, Berroir J-M, Glattli D C, Plaçais B, Fève G, Albert M, Flindt C and Büttiker M 2012 Current noise spectrum of a single-particle emitter: theory and experiment *Phys. Rev.* B **85** 165438
[28] Bocquillon E *et al* 2012 Electron quantum optics: partitioning electrons one by one *Phys. Rev. Lett.* **108** 196803
[29] Bocquillon E, Freulon V, Berroir J M, Degiovanni P, Plaçais B, Cavanna A, Jin Y and Fève G 2013 Coherence and indistinguishability of single electrons emitted by independent source *Science* **339** 1054–7
[30] Gabelli J and Reulet B 2013 Shaping a time-dependent excitation to minimize the shot noise in a tunnel junction *Phys. Rev.* B **87** 075403
[31] Dubois J, Jullien T, Portier F, Roche P, Cavanna A, Jin Y, Wegscheider W, Roulleau P and Glattli D C 2013 Minimal-excitation states for electron quantum optics using levitons *Nature* **502** 659–63
[32] Jullien T, Roulleau P, Roche B, Cavanna A, Jin Y and Glattli D C 2014 Quantum tomography of an electron *Nature* **514** 603–7
[33] Fricke L *et al* 2013 Counting statistics for electron capture in a dynamic quantum dot *Phys. Rev. Lett.* **110** 126803
[34] Giblin S P, See P, Petrie A, Janssen T J B M, Farrer I, Griffiths J P, Jones G A C, Ritchie D A and Kataoka M 2016 High-resolution error detection in the capture process of a single-electron pump *Appl. Phys. Lett.* **108** 023502
[35] Riwar R-P, Splettstoesser J and König J 2013 Zero-frequency noise in adiabatically driven interacting quantum systems *Phys. Rev.* B **87** 195407
[36] Dittmann N and Splettstoesser J 2018 Finite-frequency noise of interacting single-electron emitters: spectroscopy with higher noise harmonics *Phys. Rev.* B **98** 115414
[37] Berry M V 1984 Quantal phase factors accompanying adiabatic changes *Proc. R. Soc.* A **392** 45–57
[38] Uhlmann A 1986 Parallel transport and 'quantum holonomy' along density operators *Rep. Math. Phys.* **24** 229–40
[39] Ning C Z and Haken H 1992 The geometric phase in nonlinear dissipative systems *Mod. Phys. Lett.* B **06** 1541–68
[40] Landsberg A S 1992 Geometrical phases and symmetries in dissipative systems *Phys. Rev. Lett.* **69** 865–8
[41] Brouwer P W 1998 Scattering approach to parametric pumping *Phys. Rev.* B **58** R10135–8
[42] Avron J E, Elgart A, Graf G M and Sadun L 2001 Optimal quantum pumps *Phys. Rev. Lett.* **87** 236601
[43] Sarandy M S and Lidar D A 2005 Adiabatic approximation in open quantum systems *Phys. Rev.* A **71** 012331
[44] Sinitsyn N A and Nemenman I 2007 Universal geometric theory of mesoscopic stochastic pumps and reversible ratchets *Phys. Rev. Lett.* **99** 220408
[45] Sinitsyn N A and Nemenman I 2007 The Berry phase and the pump flux in stochastic chemical kinetics *Europhys. Lett.* **77** 58001
[46] Yoshii R and Hayakawa H 2013 Analytical expression of geometrical pumping for a quantum dot based on quantum master equation (arXiv:1312.3772)
[47] Pluecker T, Wegewijs M R and Splettstoesser J 2017 Gauge freedom in observables and Landsberg's nonadiabatic geometric phase: pumping spectroscopy of interacting open quantum systems *Phys. Rev.* B **95** 155431
[48] Pluecker T, Wegewijs M R and Splettstoesser J 2017 Meter calibration and the geometric pumping process in open quantum systems (arXiv:1711.10431)
[49] Nakajima S, Taguchi M, Kubo T and Tokura Y 2015 Interaction effect on adiabatic pump of charge and spin in quantum dot *Phys. Rev.* B **92** 195420
[50] Pekola J P, Saira O-P, Maisi V F, Kemppinen A, Möttönen M, Pashkin Y A and Averin D V 2013 Single-electron current sources: toward a refined definition of the ampere *Rev. Mod. Phys.* **85** 1421–72
[51] Battista F, Haupt F and Splettstoesser J 2014 Energy and power fluctuations in ac-driven coherent conductors *Phys. Rev.* B **90** 085418
[52] Battista F, Haupt F and Splettstoesser J 2014 Correlations between charge and energy current in ac-driven coherent conductors *J. Phys.: Conf. Ser.* **568** 052008
[53] Moskalets M 2014 Floquet scattering matrix theory of heat fluctuations in dynamical quantum conductors *Phys. Rev. Lett.* **112** 206801
Moskalets M 2014 *Phys. Rev. Lett.* **113** 069902 (erratum)
[54] Terrén Alonso P, Romero J and Arrachea L 2019 Work exchange, geometric magnetization, and fluctuation–dissipation relations in a quantum dot under adiabatic magnetoelectric driving *Phys. Rev.* B **99** 115424
[55] Popovic M, Mitchison M T, Strathearn A, Lovett B W, Goold J and Eastham P R 2020 Non-equilibrium quantum thermodynamics with time-evolving matrix product operators (arXiv:2008.06491)
[56] Bagrets D A and Nazarov Y V 2003 Full counting statistics of charge transfer in Coulomb blockade systems *Phys. Rev.* B **67** 085316
[57] Mostafazadeh A 1997 Quantum adiabatic approximation and the geometric phase *Phys. Rev.* A **55** 1653–64
[58] Wunsch B, Braun M, König J and Pfannkuche D 2005 Probing level renormalization by sequential transport through double quantum dots *Phys. Rev.* B **72** 205319
[59] Braun M, König J and Martinek J 2004 Theory of transport through quantum-dot spin valves in the weak-coupling regime *Phys. Rev.* B **70** 195345
[60] Cavaliere F, Governale M and König J 2009 Nonadiabatic pumping through interacting quantum dots *Phys. Rev. Lett.* **103** 136801
[61] Saptsov R B and Wegewijs M R 2012 Fermionic superoperators for zero-temperature nonlinear transport: real-time perturbation theory and renormalization group for Anderson quantum dots *Phys. Rev.* B **86** 235432

[62] Saptsov R B and Wegewijs M R 2014 Time-dependent quantum transport: causal superfermions, exact fermion-parity protected decay modes, and Pauli exclusion principle for mixed quantum states *Phys. Rev.* B **90** 045407
[63] Schulenborg J, Splettstoesser J, Governale M and Contreras-Pulido L D 2014 Detection of the relaxation rates of an interacting quantum dot by a capacitively coupled sensor dot *Phys. Rev.* B **89** 195305
[64] Splettstoesser J, Governale M, König J and Fazio R 2006 Adiabatic pumping through a quantum dot with coulomb interactions: a perturbation expansion in the tunnel coupling *Phys. Rev.* B **74** 085305
[65] Reckermann F, Splettstoesser J and Wegewijs M R 2010 Interaction-induced adiabatic nonlinear transport *Phys. Rev. Lett.* **104** 226803
[66] Büttiker M, Thomas H and Prêtre A 1994 Current partition in multiprobe conductors in the presence of slowly oscillating external potentials *Z. Phys. B: Condens. Matter* **94** 133–7
[67] Brouwer P W 2001 Rectification of displacement currents in an adiabatic electron pump *Phys. Rev.* B **63** 121303
[68] Avron J E, Elgart A, Graf G M and Sadun L 2004 Transport and dissipation in quantum pumps *J. Stat. Phys.* **116** 425–73
[69] Splettstoesser J, Governale M, König J and Fazio R 2005 Adiabatic pumping through interacting quantum dots *Phys. Rev. Lett.* **95** 246803
[70] Moskalets M and Büttiker M 2002 Floquet scattering theory of quantum pumps *Phys. Rev.* B **66** 205320
[71] Gasparinetti S, Viisanen K L, Saira O-P, Faivre T, Arzeo M, Meschke M and Pekola J P 2015 Fast electron thermometry for ultrasensitive Calorimetric detection *Phys. Rev. Appl.* **3** 014007
[72] Wang L B, Saira O-P and Pekola J P 2018 Fast thermometry with a proximity Josephson junction *Appl. Phys. Lett.* **112** 013105
[73] Placke B A, Pluecker T, Splettstoesser J and Wegewijs M R 2018 Attractive and driven interactions in quantum dots: mechanisms for geometric pumping *Phys. Rev.* B **98** 085307
[74] Hamo A, Benyamini A, Shapir I, Khivrich I, Waissman J, Kaasbjerg K, Oreg Y, von Oppen F and Ilani S 2016 Electron attraction mediated by Coulomb repulsion *Nature* **535** 395–400